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Monitors for Prediction of Neptunium and Americium Concentrations in Spent Fuel

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The higher-actinide isotopes of ^{237}Np, ^{241}Am, and ^{243}Am are fissionable nuclides formed in reactor fuels and, if separated, could conceivably be used in a nuclear explosive device. Recently, the International Atomic Energy Agency Board of Governors tasked the Secretariat with maintaining oversight of these Np and Am nuclides, or alternate nuclear materials (ANMs). Calculations of the production of these isotopes using reactor analysis codes may provide a cost-effective route to determine levels of these materials in spent fuel. Several codes and data libraries have been examined for their ability to accurately predict the quantity of the ANMs available from several reactor types.

Introduction

Trace quantities of neptunium (Np) are present in nature as a result of neutrons producing transmutation reactions in uranium (U) ores; larger quantities are produced in nuclear power reactors. Np is formed by beta-decay of ^{237}U, which forms after two subsequent neutron captures of ^{235}U. The main reactions leading to Np are shown in Figure 1.

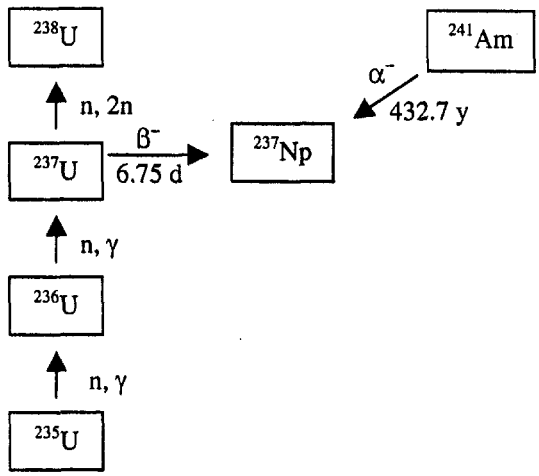

Figure 1. Buildup and decay reactions leading to Np-237.

The americium (Am) isotopes are mainly formed by beta decay of plutonium isotopes. The reactions leading up to these isotopes are shown in Figure 2. While a small amount is produced during irradiation of nuclear fuel, the largest contribution to the concentration of Am in spent fuel is from the decay of Pu-241. Both Am and Np isotopes are not consumed in most reactor types because fission of these isotopes requires fast neutrons, such as those present in breeder reactors or accelerators.

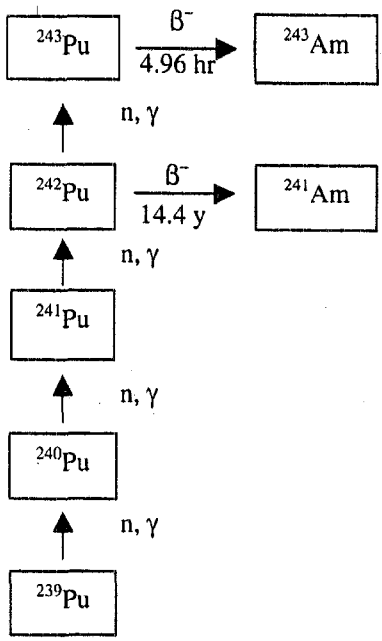

Figure 2. Buildup and decay reactions leading to the isotopes Am-241 and Am-243.

Removing Np, which has a 2.1 million year half-life, and Am-241 and Am-243, with 432.7 and 7,370 year half-lives, respectively, dramatically reduces the long-term toxicity.[1,2] Therefore, interest in recycling Np as well as Am back into the fuel cycle to decrease the radio-toxicity of disposed waste is high. Considerable research effort has gone into techniques for separating these actinides (^{237}Np, ^{241}Am, ^{243}Am) from spent fuel and a large number of methods, using chemical separation to optical techniques, are described in the literature.[3-6]

Concerns other than the radio-toxicity of these isotopes exist; in separated form, these isotopes could also be used for the construction of nuclear explosives. This possible use has been known; however, when the International Atomic Energy Agency (IAEA) originally adopted the IAEA Statue[7], these materials did not exist in quantity and were not, therefore, designated as special fissionable materials, for which safeguards were established. However, according to Article XX of the Statute, additional materials may be designated as special fissionable materials. The amount of these Am and Np nuclides or alternate nuclear materials (ANMs), has grown tremendously from nuclear power cycles. As mentioned above, effective separation technologies have been developed. In September of 1999, the IAEA Board of Governors tasked the Secretariat with oversight of these materials.[8]

The monitoring scheme[9] initiated by the IAEA does not yet require full material balance accounting, but knowledge of the amounts of these nuclides present in spent fuel is vital for determining appropriate measures needed for this oversight. Also, a cost-effective means of accounting for the materials is desirable. Calculational methods may aid in a cost-effective approach for both facility operators and the inspection organization to monitor these materials.

Reactor Physics Codes

Here, three different reactor analysis codes are examined for their ability to accurately predict the amounts of ANMs present in spent fuels from several different reactor types. The reactor analysis codes evaluated are ORIGEN2, HELIOS, and MONTEBURNS. All three codes use nuclear data derived from Evaluated Nuclear Data Files (ENDF).

ORIGEN2[10] is a burnup and depletion code that uses the exponential method to solve the time-dependent burnup equations. This code has been used widely for fuel management, shielding studies, and safeguards investigations. In this code, predetermined, reactor-specific libraries give one-group cross sections, flux spectra, and fission yields. Current ORIGEN2 libraries include ones for pressurized and boiling water reactors (PWRs and BWRs), liquid metal fast breeder reactor (LMFBR) driver and blanket fuel, and Canada Deuterium-Uranium (CANDU) reactors.

As a result of its widespread use, ORIGEN2 has undergone numerous benchmarking studies to examine the code's ability to concentrations of uranium, plutonium, and fission products in spent nuclear fuel. Some effort has been expended on determining the codes accuracy in predicting Am-241 and Am-243 concentrations.[11-14]

HELIOS[15] is a lattice physics code that uses the method of angularly dependent, current-coupled collision probabilities to solve the neutron transport equation. HELIOS-1.4 uses a set of multigroup cross sections derived from thee ENDF/B-VI nuclear data file. Although HELIOS has been benchmarked for critical experiments, assembly pin powers, and uranium, plutonium and fission product isotopics, quantification of the code's ability to predict higher-actinide concentrations is limited.

Monteburns[16] is a code system that links the Monte Carlo simulation code MCNP[17] to ORIGEN2. Monteburns has a UNIX c-shell command file that links the codes by calling a FORTRAN77 program that processes input and output files for the codes. The user inputs values to specify system geometry, initial material composition, and other parameters required by MCNP and ORIGEN2. Monteburns generates time-dependent results and compiles the output results from ORIGEN2 and MCNP into a more compressed set of files for post-processing.

ORIGEN2 differs significantly from the HELIOS and Monteburns in that ORIGEN2 does not use a transport calculation for the specific case to determine collapsed cross sections and fluxes.

Models

Using the three codes, simulation of the irradiation of fuel for the following reactor types were performed: pressurized water reactors (PWRs), boiling water reactors (BWRs), Russian water-cooled, water-moderated reactors (VVERs), pressurized heavy water reactors (PHWR), high temperature gas reactors (HTGRs). Assumed average specific power for assemblies were used. Actual reactor power histories were not available for use in the calculations. Each fuel was modeled to burnups appropriate to the reactor type, using 1000 MWd/MTU burnup steps. For HELIOS-1.4 and Monteburns calculations, a two-dimensional pin cell model was used.[18]

Results

Code accuracy in determining all three isotopes is evaluated with respect to pressurized water reactors (PWRs), boiling water reactors (BWRs), Russian water-cooled, water-moderated reactors (VVERs), pressurized heavy water reactors (PHWR), and high temperature gas reactors (HTGRs). Radiochemical measurements of the higher actinides from the literature were used to judge code accuracy.

The average percent differences between calculated and measured values for Np and Am concentrations are given in Tables 1-3 below. Accuracy of the value of isotope concentration varies with the code used and type of reactor examined.

Table 1. Comparison of average percent difference between calculated and measured values of Am-241 for different reactor types and reactor physics code used

Reactor type	HELIOS	ORIGEN2	Monteburns
PWR	3.7	4.6	4.1
BWR	12.1	12.2	12.2
VVER-440	7.3	32.4	10.5
PHWR	10.1	14.3	11.2
HTGR	5.8	6.2	8.8

Table 2. Comparison of average percent difference between calculated and measured values of Am-243 for different reactor types and reactor physics code used

Reactor type	HELIOS	ORIGEN2	Monteburns
PWR	17.7	14.4	16.8
BWR	13.6	17.0	20.9
VVER-440	9.8	139.9	11.6
PHWR	14.2	18.2	12.2
HTGR	8.7	15.1	13.2

Table 3. Comparison of average percent difference between calculated and measured values of Np-237 for different reactor types and reactor physics code used

Reactor type	HELIOS	ORIGEN2	Monteburns
PWR	8.2	4.3	6.2
BWR	7.3	5.6	3.2
VVER-440	7.9	43.2	5.8
PHWR	11.2	15.6	1.3
HTGR	7.6	9.2	4.3

The amount of Am-241 in spent fuel is highly dependent on the decay time of the fuel; the decay of Pu-241 is the main contributor to this isotope's concentration in spent fuel (see Figure 3).

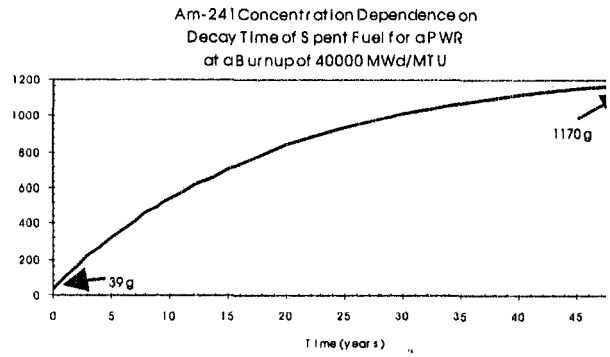

Figure 3. The concentration of Am-241 has a strong dependence on decay time of the spent fuel, as a result of the decay of Pu-241.

Discussion of results

The ORIGEN2 code did well for BWR and PWR fuel. However, the ORIGEN2 pressurized water reactor libraries are not sufficient for prediction of ANMs in spent fuel from VVER-440 reactors. Both HELIOS and Monteburns codes were able to calculate these isotopes well. Also, scatter exists in the experimental radiochemical measurements of these isotopes, so accuracies may be insufficient for safeguard purposes.

Another important point to note is that HELIOS does not include alpha decay of Am-241 to Np-237.

Few measurements of the higher-actinide concentrations in spent fuel have been published, and additional information in this arena would be helpful for further analysis. For some reactor types, modification of existing data libraries may be necessary for cost-effective oversight of ANMs using calculational methods. Furthermore, specific libraries for VVER, RBMK, and BN-350 would be useful.

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